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On the inclusion of Limb Infrared Monitor of the Stratosphere version 6 ozone in a data assimilation system

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[1] Version 6 ozone profiles for 1978–1979 from the Limb Infrared Monitor of the Stratosphere experiment on the NIMBUS 7 satellite (or LIMS v6) are assimilated into an updated version of the GEOS-5 model of NASA. First, an assimilation study is carried out using GEOS-5 version 7.2 (v7.2) and solar backscatter ultraviolet (SBUV) version 8.6 ozone profiles. Then, a second study is conducted that ingests both the LIMS and SBUV ozone, as weighted by their estimated absolute error vectors. Ozone from this second study compares well with independent observations from the Stratospheric Aerosol and Gas Experiment (SAGE I) and from the time series of ozonesonde data at Hohenpeissenberg and at Wallops Island. Assimilation of the LIMS data gives improved ozone distributions in the upper stratosphere (pressure < 5 hPa) and in the polar night region—the latter where solar backscatter ultraviolet (SBUV) is not observed. The LIMS ozone leads to improved total column ozone analyses in winter/spring outside of the tropics, based on independent comparisons with total ozone from the Total Ozone Mapping Spectrometer. The LIMS ozone also adds information in the tropics on coherent structural features at 20–30 hPa, related to the phase transition of the quasi-biennial oscillation wind field. It is affirmed that the process of data assimilation represents a cost-effective way of characterizing new and/or reprocessed satellite ozone data sets. It is concluded that the GEOS-5 v7.2 model with the addition of the LIMS data can improve analyses of ozone in 1978–1979.

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1. Introduction

[2] A key goal for the Earth Science Program of NASA is to provide a long-term, continuous characterization of the Earth system for addressing weather, atmospheric composition, and climate change issues. In pursuit of this goal, the Modern-Era Retrospective Analysis for Research and Applications (MERRA) system [*Rienecker et al.*, 2008, 2011] has been using data assimilation techniques to integrate millions of in situ and satellite observations with model forecasts, as part of the Goddard Earth Observing System, Version 5, data assimilation system (GEOS-5 DAS). Global analyses have been archived from 1 January 1979 to the present day.

[3] Atmospheric ozone is a key, radiatively active component in the climate system. Improved characterization of its global distribution, temporal evolution, or trends is critical to understanding and predicting changes in chemical composition and climate. *Stajner et al.* [2001] assimilated partial column ozone from the solar backscatter ultraviolet (SBUV) instrument—a nadir-viewing radiometer—along with total column ozone from the Total Ozone Mapping Spectrometer (TOMS) into an early version of the GEOS model with good success. SBUV measures backscattered radiation at 12 discrete wavelengths from 255 to 340 nm; its ozone profiles are limited to sunlit regions [*Bhartia et al.*, 1984; *McPeters et al.*, 1984]. The vertical resolution of an SBUV ozone profile is estimated to be 6–8 km from the upper to middle stratosphere, respectively, but then degrades to only 10–15 km in the lower stratosphere [*Bhartia et al.*, 2012]. Currently, atmospheric reanalyses are being carried out via assimilation of ozone data from a succession of SBUV instruments for MERRA and for the interim version of the next ECMWF Reanalysis (ERA-Interim) [*Dee et al.*, 2011].

[4] Satellite, limb-emission instruments provide stratospheric species profile data both day and night and with improved

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vertical resolutions. As an example, *Ménard and Chang* [2000] characterized the covariance parameters for the observed and forecast tracer-like species CH_4 and showed that one can gain improved forecasts in the lower stratosphere from the assimilation of its profiles from a limb-viewing experiment. Ozone also serves as a tracer-like species for the lower stratosphere; however, its distribution and spatial gradients differ from those of CH_4, especially in the vertical. To determine the value of ozone from limb measurements, *Wargan et al*. [2005] first conducted a Control assimilation experiment that included both the SBUV partial column and total ozone data. In a second experiment, they added limb-infrared ozone profiles from the Michelson Interferometer for Passive Atmospheric Sounding (MIPAS) satellite experiment and as characterized by *Migliorini et al*. [2004], and they found substantial impacts and forecast gains from the inclusion of its ozone during the Northern Hemisphere winter of 2002/2003. *Geer et al*. [2006] and *Errera et al*. [2008] reported similar gains for a range of ozone data assimilation systems that employed MIPAS ozone for the longer period of mid-2002 through early 2004. More recently, *Stajner et al*. [2008] and *Wargan et al*. [2010] have had success from assimilating ozone data from the Microwave Limb Sounder (MLS) plus the Ozone Monitoring Instrument of the EOS AURA satellite, in order to better characterize variations in the ozone distributions of both the troposphere and lower stratosphere.

[5] The Limb Infrared Monitor of the Stratosphere (LIMS) instrument operated from NIMBUS-7 and provided an additional, complementary source of limb ozone data to that from SBUV for assimilation in the earlier period of 1978–1979. Unlike SBUV, the infrared limb emission measured by LIMS provided daily ozone profiles from 64°S to 84°N and at two local times per latitude, but only from late October 1978 through late May 1979 [*Gille and Russell*, 1984]. *Austin* [1992] was the first to use the species profiles from LIMS to determine the feasibility of their assimilation into a chemical transport model (CTM) of the stratosphere, and he reported that the species distributions from the model agreed with the data in most instances. An updated, LIMS version 6 (v6) ozone data set is now available and extends from cloud top and into the upper mesosphere with a vertical resolution of 3.7 km [*Remsberg et al*., 2004]. LIMS also obtained data beyond the reach of SBUV through the polar night of the high northern latitudes. Consequently, LIMS offers the potential for improving the ozone analyses in the polar winter.

[6] *Remsberg et al*. [2007] included results of comparisons with several independent data sets for the purpose of validating the LIMS v6 ozone. In particular, they showed zonally averaged differences for LIMS versus SBUV version 8 (or v8) ozone for three representative days across the 7 month span of LIMS observations. They reported that the two data sets agreed within their combined estimates of error, although they also found coherent vertical structures in the plots of their zonally averaged differences that varied slightly from November to May. Data assimilation studies can further support an evaluation of the quality of the ozone for the LIMS time period, when there were relatively few, correlative measurements [e.g., *Lahoz et al*., 2007]. As was found for the MIPAS and MLS ozone, we will show that the LIMS profiles resolve more of the vertical ozone structure and provide improved ozone analyses.

[7] Section 2 describes the model used in this study, GEOS-5 version 7.2, which is an update from the version used in MERRA. We conducted two assimilation experiments, a Control case based on ozone from SBUV and a LIMS analysis case using the ozone from both SBUV and LIMS. Section 3 describes the monthly averaged differences for the ozone between the LIMS analysis and Control cases. Section 3 also shows mean and root-mean-square (RMS) comparisons with independent ozone profile measurements from the Stratospheric Aerosol and Gas Experiment (SAGE I), from time series of ozonesonde soundings at Hohenpeissenberg and at Wallops Island, and from comparisons against total column ozone obtained with the TOMS experiment on Nimbus 7. Section 4 concludes that the LIMS v6 ozone is of good quality and that its assimilation into GEOS-5 provides improved analyses of the ozone distributions for many regions of the stratosphere.

2. GEOS-5 and the Assimilation of LIMS Ozone

2.1. Preliminary Studies

[8] The GEOS-5 general circulation model transport is based on finite-volume dynamics [*Lin*, 2004] that was shown to be important for maintaining the integrity of the ozone field in forecasts [*Pawson et al*., 2007]. The data assimilation system (DAS) is based on a Grid-point Statistical Interpolation (GSI) scheme [*Wu et al*., 2002; *Purser et al*., 2003a, 2003b] having a 6 h update cycle. Although ozone is one of the main state vectors, there are no cross-correlation terms within the GSI package connecting the ozone to the other meteorological variables. For the long-term MERRA simulations, the GEOS-5 model transports the odd oxygen family and approximates ozone photochemistry using prescribed zonal mean, monthly averaged, ozone production and loss frequencies obtained from a 2-D CTM, whose chemical equilibrium states are based on the first 7 years of data from the MLS and Halogen Occultation Experiment instruments of the Upper Atmosphere Research Satellite or for the mid-1990s.

[9] *Stajner et al*. [2001, 2004] and *Struthers et al*. [2002] showed that the output of the ozone DAS can be used to characterize and verify estimates of the error covariances of both the forecast model and the satellite ozone observations. Specifically, the GSI package calculates normalized values for the observation (O) minus forecast (F) results, or $[0.5 \times (O - F)^2/(\sigma^2_{obs})]$, where σ^2_{obs} is the SBUV observation error variance. A so-called "cost function" for the ozone is then obtained by dividing the foregoing quantity by the number of observations—a χ^2 type of statistic; the optimum χ^2 value is 1.0 [*Ménard et al*., 2000]. Nevertheless, a value as large as 2.0 may still imply that the characterization of the combined error is reasonable, given that there are model/data biases and that one only has an approximate knowledge of the model/background errors.

[10] Initially, we conducted a Control case with MERRA, using all observations from 1 November of the MERRA observing system, including SBUV Version 8.0 (v8.0) partial column ozone data. The cost function for that case was near 2.5, indicating some imbalance in the system perhaps from an underestimate of the SBUV errors. In contrast, *Stajner*

Table 1. Assigned Absolute Error Estimates for SBUV v8.6 Ozone (in DU) as a Function of Pressure-Layer for the Mesosphere and Stratosphere

Pressure at Bottom of Layer (hPa)	Estimated Ozone Error (in DU, 1-sigma)
0.100	1.000
0.158	1.000
0.251	1.000
0.398	1.000
0.631	1.000
1.000	1.000
1.580	1.000
2.510	1.000
3.980	1.021
6.310	1.265
10.00	1.358
15.80	1.225
25.10	1.225
39.80	1.423
63.10	1.871
100.0	1.871
158.0	1.831
251.0	1.730

et al. [2001] achieved a χ^2 value of 1.52, but their SBUV error covariance matrix included off-diagonal terms to account for the significant correlations between adjacent layers; the GEOS-5/GSI code does not support off-diagonal terms. In other words, the observation operator within the current ozone DAS assumes that the SBUV ozone layers are independent and/or have flat vertical-weighting functions, even though that is not the case.

[11] A preliminary LIMS analysis case was also carried out with MERRA. It used the same set of SBUV observations as the Control, but with the addition of LIMS v6 ozone profiles for November 1978 through May 1979 (~450,000 profiles). However, by January, the output files showed that ozone values below about the 40 hPa level had become much smaller than the values from either the SBUV or the LIMS profiles that had been assimilated. It was determined that the decrease in ozone was due to the vertical advection from near the 70 hPa level of LIMS ozone that is characterized by two modes—a nominal mode near 0.5 parts per million by volume (ppmv) and a primary mode at about 0.05 ppmv. The latter mode is a consequence of retrieving to a LIMS ozone value equivalent to the radiance noise of the LIMS ozone channel. The mode at 0.05 ppmv also leads to a distribution of LIMS analysis O – F residuals that are non-Gaussian and thus do not meet an important assumption for the data assimilation algorithm (see Appendix A for details). That problem was corrected to first order by screening out LIMS ozone values of < 0.1 ppmv below the 50 hPa level, prior to their assimilation.

[12] Are the χ^2 values improved from the assimilation of the LIMS versus the SBUV data? To address this point, we then conducted an assimilation study for November and December by ingesting only the LIMS v6 ozone profiles from 25 October 1978 onward and then introducing the SBUV partial column ozone on 1 November 1978 and thereafter. Time series of the cost function from the inclusion of just the LIMS ozone dropped from an initial value of about 4.0 to a value near 2.0 in 2 days—a rapid adjustment. The addition of the SBUV data from 1 November onward gave a χ^2 value slightly smaller than 2.0 or not much additional improvement. It is noted though that this value of 2.0 is smaller than the value of 2.5 obtained from our longer SBUV Control study, because the latter case extended through Northern Hemisphere winter/spring, a time of more vigorous transport and mixing that is more difficult to model. The expectation from these several preliminary studies was that the assimilation of the LIMS ozone ought to improve the ozone analyses for the stratosphere.

2.2. Final Control and LIMS Analysis Experiments

[13] A number of changes were made for our final LIMS analysis studies, focusing on 1978–1979 and in light of the findings from the preliminary LIMS assimilation experiments. First, an updated GEOS-5 DAS (version 7.2) was used (http://gmao.gsfc.nasa.gov/products/). Its primary updates pertain to the characterization of clouds and precipitation in the troposphere and have no bearing on the results of these final Control and LIMS analysis cases. Gravity wave drag and radiation schemes are unchanged for the stratosphere. In this version, radiances from the Stratospheric Sounding Unit are now ingested into GSI through a Community Radiative Transfer Model [*Chen et al*., 2011], which is a substantial advance from MERRA [*Rienecker et al*., 2011]. The experiments in this study used a lower, latitude by longitude, spatial resolution (or $1° \times 1.25°$) than MERRA, but retained the same 72 layers. Ozone was initialized using the MERRA field from 28 September 1979, since MERRA began in 1979. In the assimilation system, the ozone spins up quickly or in 10 days.

[14] *Coy et al*. [2007] found a reduction in O – F standard deviations in their model studies, when they adopted a reference state for the chemical module that was specific to the observation year of their assimilated ozone. Therefore, new ozone production and loss rates were substituted into the chemistry module for the current studies, based on an offline, time-dependent 2-D model that uses chlorine source gas amounts appropriate for the stratosphere of 1978 to 1979 [*Natarajan et al*., 2002]. This revised chemistry module is used for both the Control (SBUV) and the LIMS analysis (LIMS plus SBUV) experiments.

[15] The observational, partial column SBUV data are based on its most recent Version 8.6 (v8.6) data set and are provided at 21 layers, even though their degrees of freedom are fewer than that because of the correlations with adjacent layers. The observational SBUV v8.6 error estimates (in Dobson units or DU) adopted for this study are given as the error vector in Table 1. These estimates are similar to those used for its monthly zonal mean partial columns [*Bhartia et al*., 2012]. As before, they do not explicitly include the effects of the lower vertical resolutions for the SBUV partial columns via off-diagonal terms from the more exact covariance matrix formulation. On the other hand, the effects of that vertical smoothing are partly accounted for because the percentage error due to the absolute error vector in Table 1 is larger in the tropics than at the middle latitudes for the lower stratosphere, as indicated in *Bhartia et al*. [2012].

[16] The LIMS v6 ozone error vector (in ppmv) is given in Table 2 and is based on a combination of its random and systematic components, but without the effects of any temperature bias. These combined LIMS errors vary from 7% at 3 hPa to 28% at 100 hPa, as referenced to the zonal

Table 2. Assigned Absolute Error Estimates for LIMS v6 Ozone as a Function of Pressure-Altitude[a]

Pressure (hPa)	Estimated Ozone Error (in ppmv, 1-sigma)
0.1	0.28
0.4	0.18
1.0	0.21
3.0	0.35
5.0	0.56
7.0	0.69
10.0	0.76
30.0	0.52
50.0	0.29
70.0	0.22
100.0	0.08

[a]Error estimates are linearly interpolated in log-pressure between levels.

mean ozone at 30°N and for a vertical resolution of 3.7 km [*Remsberg et al*., 2007]. The LIMS absolute error vector also leads to percentage errors that are on the order of those from SBUV for most of the stratosphere. An exception is for the uppermost stratosphere and the lower mesosphere, where the percentage LIMS errors are smaller than from SBUV.

[17] In general, the DAS calculates the analysis state by taking into account both the observation errors (assumed to be uncorrelated in the vertical) and the background error covariances. The present DAS version assumes that the background error variance is 20% versus 5%, respectively, of the background ozone in the troposphere and the stratosphere, where background ozone is in terms of kg/kg. Thus, the background errors are state-dependent. This approach to background error modeling is empirical rather than derived from first principles, but it has been extensively tested and shown to be useful [e.g., *Stajner et al*., 2008; *Wargan et al*., 2010]. The underlying idea is that the background errors (and therefore analysis increments) should be larger (smaller) where ozone is larger (smaller) in order to resolve sharp gradients across air mass boundaries, such as the tropopause. In the context of the assimilation of the SBUV data in GEOS-5, it was found that the smaller-scale, low ozone laminae of the upper troposphere/lower stratosphere are resolved much better with this approach than with static background errors. The DAS also discards observations for which O − F values exceed a certain threshold. That quality control threshold varies from 1 to 7 DU, depending on layer (e.g., 7 DU at 40–25 hPa). LIMS observations are discarded if the ratio $|O - F|/[\text{obs error}] > 10$.

[18] From the LIMS analysis experiment, we calculated the two separate, SBUV and LIMS, components of the cost function, based on their respective values of $(O - F)^2$ and σ^2_{obs}. Average stratospheric cost functions for the 5 months of 1979 are 2.56 for SBUV and 2.32 for LIMS. These cost functions are larger than optimal, in part, because both the vertical averaging for SBUV and the horizontal averaging along the tangent path for the LIMS profiles extend across several model layers or grid boxes, respectively. Winds from GEOS-5 transport the ozone fields at each 6 h time step and generate small-scale spatial structure that is not resolved well with the data. The calculated differences, or the O − F residuals of the cost function, are affected by the smaller-scale structure of the forecast ozone field. It is also more difficult to forecast structure in the lower stratosphere for a tracer like ozone as opposed to methane because of the much larger vertical gradients of the ozone profiles.

3. Impact of Assimilating LIMS v6 Ozone

[19] The current Control (SBUV) and LIMS analysis (LIMS + SBUV) results that follow were obtained using GEOS-5.7.2 and the ozone DAS described in section 2. The model for both runs was spun up for 4 weeks before assimilation of any ozone data. In section 3.1, we present examples of the differences for monthly, zonally averaged ozone distributions from the assimilations. Then, we evaluate the results from the two runs in sections 3.2 and 3.3 using the independent ozone data sets of SAGE I and from ozonesondes. Section 3.4 shows differences for total column ozone fields from the two cases for both January and April and then compares those results with the TOMS total ozone distributions.

3.1. Monthly, Zonally Averaged Ozone Distributions

[20] First, we show in Figures 1a and 1b the ozone mixing ratio distributions for April 1979 from the LIMS v6 Level 2 or profile data-set and from the SAGE I v6.1 profile data-set. The LIMS ozone distributions extend from 84°N to 64°S for each of its 7 months. The SAGE I instrument began making measurements in late February 1979, so it provides ozone comparison data for LIMS from late February through May [*McCormick et al*., 1989]. The occultation measurements of SAGE I for April were between 50°N and 60°S, which is the span of tangent point latitudes of its sunrise and sunset locations. One can see that the LIMS and SAGE I zonal mean ozone distributions appear very similar with latitude and altitude. It is noted that the SAGE I ozone profile data shown here and used later in section 3.2 have not been adjusted upward in altitude by the 200 m (tropics) to 400 m (middle latitudes) developed by *Wang et al*. [1996] in part because of uncertainties about the use of the solar ephemeris data in the SAGE algorithms. There are also several anomalously large values of the LIMS zonal mean ozone in Figure 1a near 70 hPa at 35°N and 40°S that are due to spuriously large values from only two profiles in the archived level 2 data; however, they have no effect on the assimilated LIMS results because of the quality controls used in the DAS.

[21] Figures 2a and 2b show the monthly mean and zonal mean ozone for April 1979 (at top) from the assimilation of the SBUV data in the Control case and (at bottom) from the LIMS analysis case. The SBUV data already provide a good representation of the principal features of the zonal mean ozone distribution, as indicated by comparing with the independent satellite data sets of Figure 1. The Control case yields a peak ozone of 10–11 ppmv in the tropical middle stratosphere with values decreasing poleward. Addition of the LIMS data for the analysis case yields an ozone distribution very similar to that of the Control, after the excessively low LIMS ozone values (< 0.1 ppmv) from below the 50 hPa level were screened out (see Appendix A).

[22] Figure 3 shows the zonal mean ozone differences for April from the values in the LIMS analysis minus the Control assimilation plots of Figure 2. Largest differences in Figure 3 are in the upper stratosphere, and there is an equator-to-pole gradient for them. At low latitudes, those

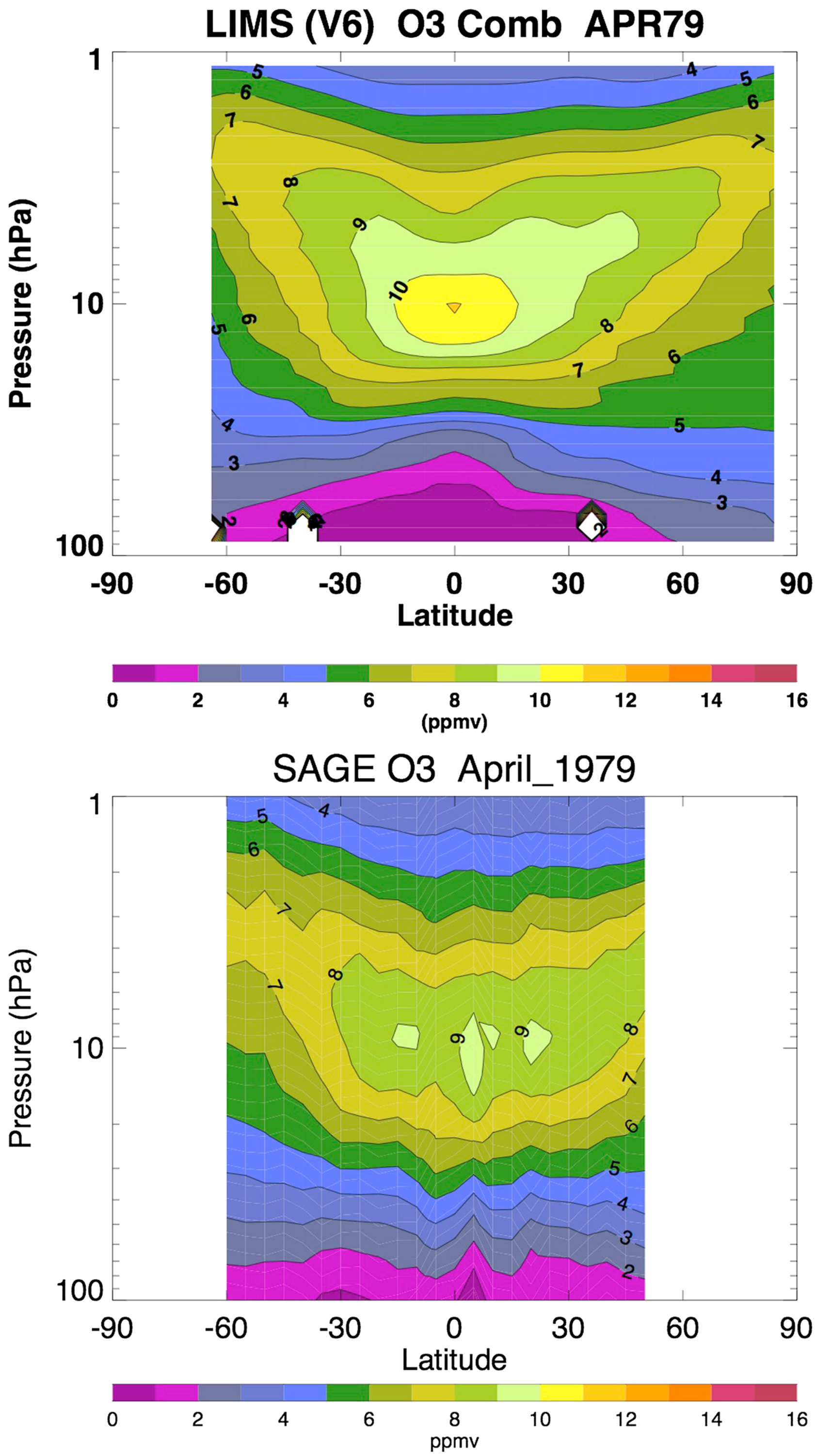

Figure 1. (a) Monthly mean, zonal mean ozone mixing ratio plots for April 1979 from the LIMS v6 Level 2 or profile data set and (b) from the SAGE I data set. Contour increment is 1.0 ppmv. Low ozone (purple/magenta); high ozone (yellow/orange).

differences are only about 0.4 ppmv or on the order of 5% to 10%, which is within the combined estimates of error for the SBUV and LIMS ozone. Odd oxygen of both the Control and the LIMS analysis cases relaxes toward chemical equilibrium rather quickly at low latitudes. Since the chemical module uses diurnally averaged production and loss frequencies, the lifetime for odd oxygen at 1 to 3 hPa is 6 to 12 h at low solar zenith angles (at the low latitudes) or on the order of the 6 h time step for the assimilation [see also *Geer et al.* 2007, Figure 1]. The LIMS ozone profiles show day minus night differences on the order of 5% in the mid to upper stratosphere [*Remsberg et al.*, 2007]. Thus, about half of

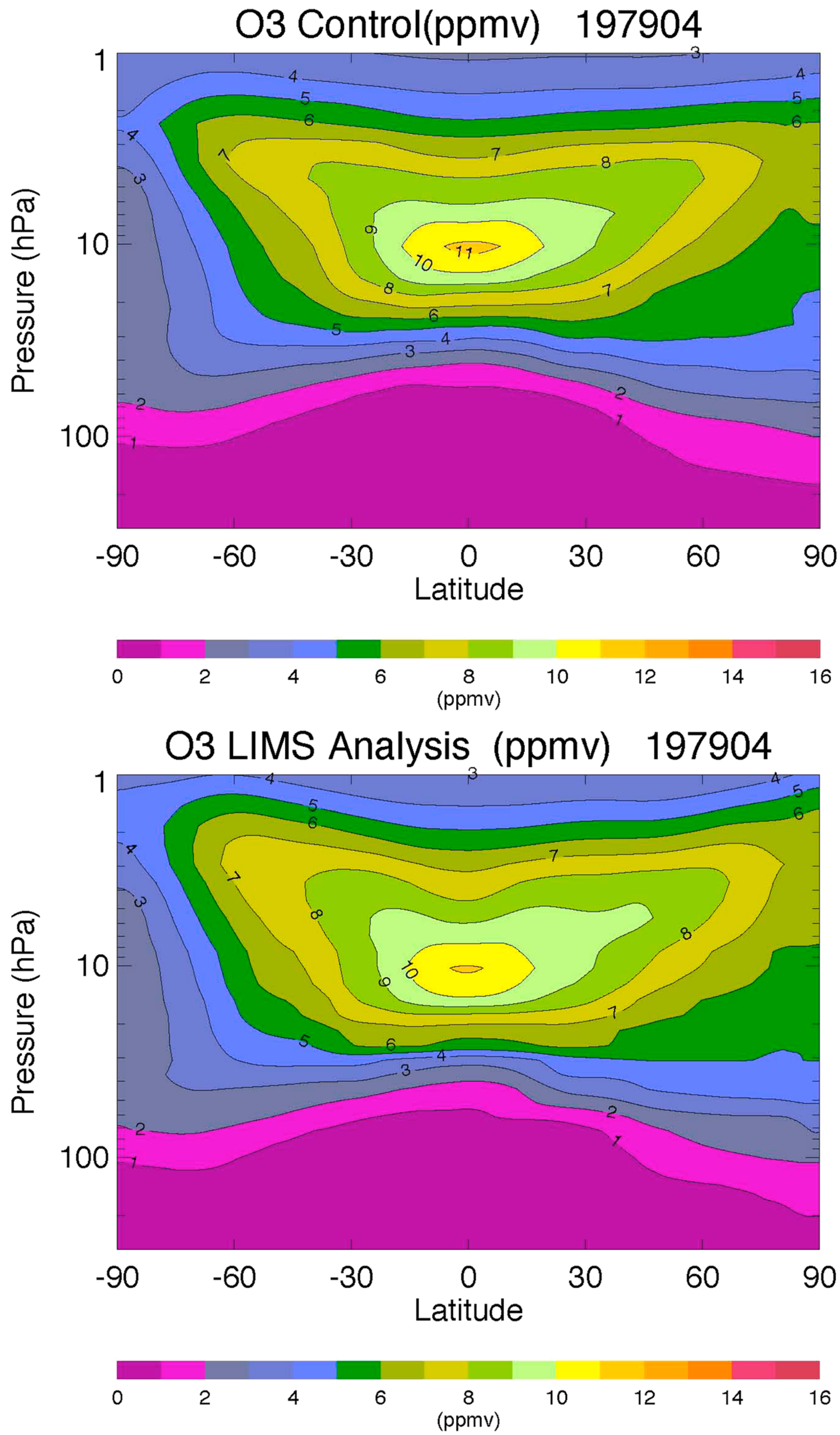

Figure 2. Ozone distributions for April 1979 (a) from the Control assimilation and (b) from the combined SBUV/LIMS analyses. Contour increment is 1.0 ppmv.

the 0.4 ppmv difference in Figure 3 is because LIMS obtained measurements both at midday and at night, while SBUV provided data only during daytime.

[23] At high latitudes, the chemical lifetime is about 1 day in the upper stratosphere [*Geer et al.*, 2007], where the ozone differences in Figure 3 are larger and on the order of 1.0 ppmv or 20% to 25% of the average ozone. At about 2 hPa, the error vector for LIMS v6 translates to a value of about 8%, while for SBUV v8.6, it is assigned a value of 1 DU or about 20%. In this instance, the results of the LIMS analysis case are weighted toward the LIMS rather than the SBUV observations, leading to the pattern of increasing differences with latitude. While the diurnally averaged ozone production and loss coefficients are the same in the Control and LIMS analysis experiments, the relative impact of the photochemistry is greater in the Control case because of the differences in error vectors. In addition, photochemical models yield an ozone deficit on the order of 10% compared to observations in this region of the

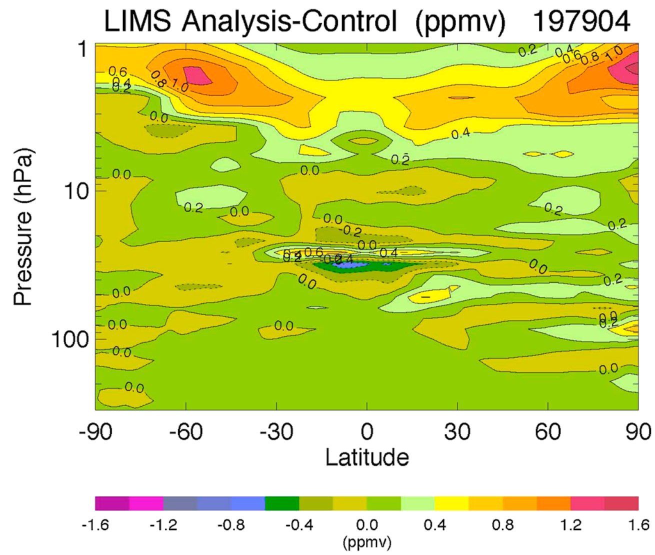

Figure 3. Zonal mean ozone mixing ratio differences for April 1979 from the LIMS analysis minus the Control. Contour increment is 0.2 ppmv. Positive differences (yellow/red); negative differences (green/blue).

stratosphere [*Natarajan et al*., 2002]. Due to the fact that assimilation of LIMS day and night data occurs more frequently than SBUV day data, the ozone in the Control experiment relaxes to slightly lower ozone mixing ratios. This effect also contributes to the positive biases from the LIMS analysis case. LIMS did not view poleward of 64°S. Therefore, the differences in Figure 3 at even higher southern latitudes are due to the SBUV/LIMS differences in ozone at 64°S plus any effects of the net poleward transport.

[24] For most regions of the middle and lower stratosphere, the differences in Figure 3 are only on the order of 0.2 ppmv and are well within the uncertainties of the SBUV and LIMS data. In other words, the Control case is providing reasonable monthly averaged results at the pressure altitudes where transport dominates the ozone, even though the vertical resolution of the SBUV data is relatively low. However, coherent vertical structure is apparent in Figure 3, indicating that the LIMS profiles are providing some additional information on small but persistent vertical variations in the ozone fields that are not resolved by SBUV. The oscillatory vertical structure at low latitudes near 30 hPa in Figure 3 is a result of the smoothing from SBUV [*Kramarova et al*., 2013]; it occurs near the zero line of the phase transition for the quasi-biennial oscillation (QBO) winds. The LIMS algorithm is sensitive to this tropical QBO feature, in part, because it employs for its ozone forward model, the LIMS-retrieved temperature (T) profiles that have the same vertical resolution as the retrieved ozone. Measured limb radiances were sampled every 0.375 km, so the effective resolutions for T and ozone are only limited by the measurement noise and by an inability to account fully for the instrument field-of-view functions of the LIMS CO_2 and ozone channels. Thus, LIMS should be resolving sinusoidal oscillations in ozone having a vertical wavelength of 5 km or more and an amplitude perturbation of 15% to 20% [*Remsberg et al*., 1984]. A very similar oscillation in tropical ozone was found by *Gray and Ruth* [1992] as part of their simulated response to the temperature variations associated with the QBO phase transition.

[25] Figure 4 shows the results from LIMS analysis minus Control for November 1978. The LIMS analysis case indicates that the ozone is greater by more than 1.0 ppmv in the upper stratosphere at two latitudes—at 50°N and at the pole. Note that SBUV provides no ozone information in November for either the Control or LIMS analysis cases in the polar night region or poleward of about 70°N. In November, the vortex was centered on the pole, and there was very little transport of SBUV ozone into the polar night region. Ozone differences at the pole are strictly because of the assimilation of the LIMS ozone. No significant changes are found in the upper stratosphere of the Southern Hemisphere, in accord with both the shorter relaxation times for ozone at those latitudes and with the general lack of planetary wave activity in summer.

[26] There is a region of negative ozone differences at high northern latitudes in the lower stratosphere. That feature is also a result of the assimilation of relatively low, LIMS-observed ozone values within the polar vortex itself [see also *Remsberg et al*., 2007, Figure 8a]. At low latitudes and extending to northern latitudes of the middle stratosphere, the ozone differences in Figure 4 are negative by as much as −0.6 ppmv—a feature that is not apparent in April (Figure 3). Seasonal differences at low latitudes are reflective of the slight differences between the actual LIMS and SBUV profiles [*Remsberg et al*., 2007], and perhaps due to the fact that the SBUV and the LIMS ozone error vectors do not change seasonally. It is more likely that LIMS is resolving some vertical structure that is smoothed in the SBUV partial column ozone near 10 hPa.

[27] Figure 5 shows the differences for December, and they are similar to those for November at most latitudes. The region of maximum positive difference near 2 hPa has shifted from about 50°N to 40°N and is related to the longer relaxation time constant for the odd oxygen chemistry at the higher latitudes during winter solstice. Notably, the ozone differences for December have decreased to near zero at the highest latitudes and are unlike those for November, even

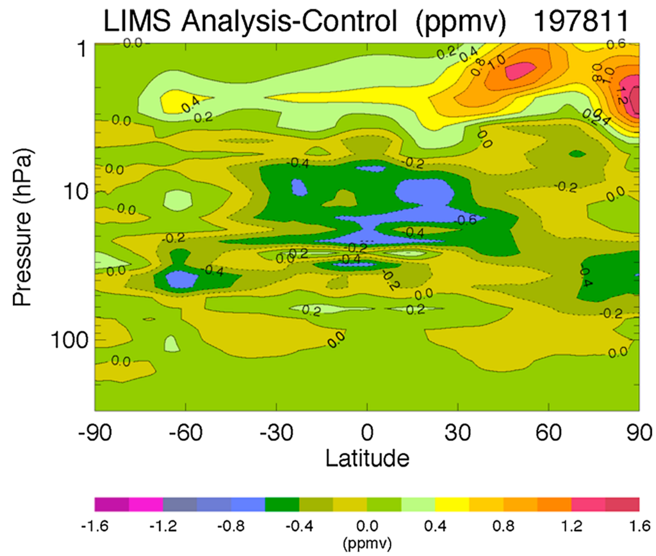

Figure 4. As in Figure 3 but for November 1978.

though there is still no SBUV ozone being assimilated in that region for the Control or the LIMS analysis cases. That change in the differences is a result of a zonal wave number 1, a "Canadian warming" event that occurred during the first week of December 1979 [*Leovy et al*., 1985]. The associated wind fields from the GEOS-5 model have led to a meridional exchange of the air in the upper stratosphere, and the SBUV ozone was transported effectively to polar latitudes in both the Control and LIMS analysis cases.

[28] Figure 6 shows the average differences between the two cases for January 1979. Once again, largest differences occur near 40°N at 2 hPa. One can easily observe the latitudinal progression of the difference maximum in the Northern Hemisphere and an increase in the differences in the Southern Hemisphere by comparing the results for November through April. The negative ozone differences in the tropics from about 20 to 7 hPa are slightly larger in January than in November and December, but recover by April. At the high northern latitudes, the ozone differences in the lower stratosphere have changed in sign from December to January. The assimilation of the LIMS data during January led to an ozone increase of about 0.2 ppmv from about 50 to 100 hPa but to an ozone decrease of the same amount from 3 to 10 hPa. This pattern of change is a result of the vigorous mixing of the air associated with a major stratospheric warming and an erosion of the polar vortex during late January 1979 [*Leovy et al*., 1985]. It is concluded that the LIMS profiles are providing more information about the transport of ozone in the northern polar region at that time.

[29] The LIMS observations minus analysis (O − A) residuals are smaller than O − F residuals, as expected from the DAS, and imply that the GEOS data assimilation system is drawing to the LIMS observations. As part of the

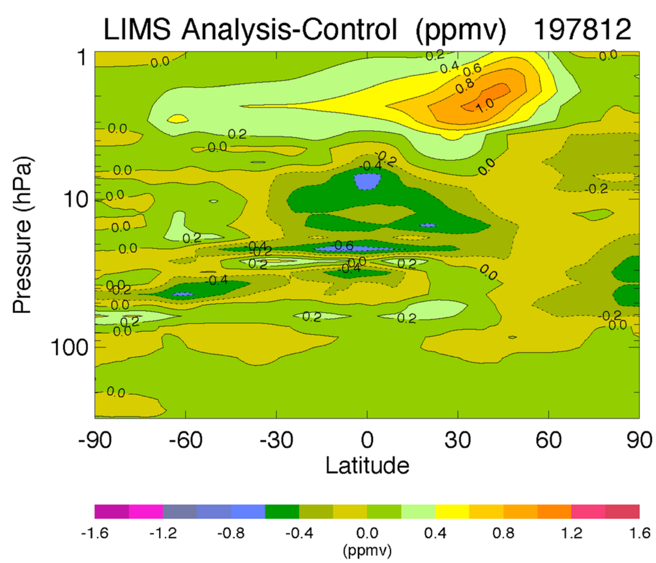

Figure 5. As in Figure 3 but for December 1978.

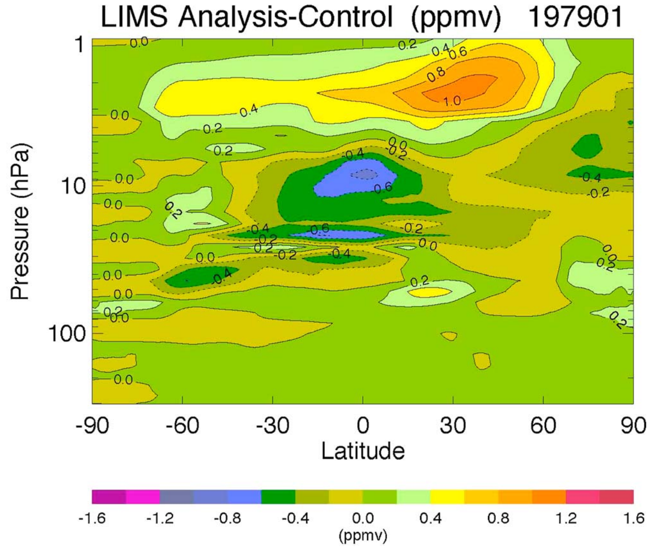

Figure 6. As in Figure 3 but for January 1979.

assimilation cycle, gridded ozone forecast and analysis fields are interpolated to the LIMS observation locations and archived every 6 h from the assimilation along with the O − F and O − A residuals. Mean O − F and O − A, or bias values are calculated for pressure levels and latitude regions, and standard deviation (SD) statistics are then obtained for them after subtracting their means. Figure 7 shows the profiles of the SDs for O − F (solid black) and O − A (dashed), and the quantity $< O^2 >$ (dotted) is the SD of the LIMS ozone itself for comparison. The SD information for SBUV or $O_{LIMS} - F_{ctrl}$ (gray) is also shown; note that we did not conduct a separate final LIMS control experiment. The analogous SD for $O_{LIMS} - A_{ctrl}$ is not shown but is very similar to $O_{LIMS} - F_{ctrl}$, in part because the SBUV and LIMS observations do not overlap by much, so there is not much ozone change from forecast to analysis at the LIMS observation locations. Figure 7a is for the latitude zone of 50°N–70°N and for January/February 1979. It shows the rather large variations that occurred due to the effects of the vigorous transport and mixing in the middle and upper stratosphere during those 2 months. The O − F (solid) has lower values than ($O_{LIMS} - F_{ctrl}$) above the 30 hPa level, indicating that the assimilation of the LIMS ozone has improved the ozone forecast. However, for most of the stratosphere, the addition of LIMS ozone does not impact the forecast by much. This finding implies that the planetary wave forecasting is captured well by the model advection and is transporting correctly the ozone that was specified from SBUV alone.

[30] Figure 7b shows the SD values for December 1978 from 70°N to 90°N or for the high latitudes during polar night. Generally, all SD values are smaller in this instance because wave activity was relatively weak, and the vortex remained centered on the pole for most of December. $O_{LIMS} - F_{ctrl}$ is exactly the same as $O_{LIMS} - A_{ctrl}$ (not plotted) in the polar cap region because no observations from SBUV were modifying ozone at polar latitudes. O − F (solid) is also smaller than $O_{LIMS} - F_{ctrl}$ (gray) throughout the stratosphere in this region, indicating that the addition of the LIMS data provides an improved ozone forecast.

[31] To summarize, the reduction of the O − F SD compared with the O SD shows forecast skill in predicting ozone variability in the Northern Hemisphere winter due in part to the quality of the previous ozone analysis that provided the initial ozone conditions for the forecast. GEOS analyses were drawn closely to the LIMS ozone profiles throughout the stratosphere, giving a near-zero bias profile (not shown). The O − A SD values are smaller than those of O − F by about a factor of two, even in the lower stratosphere. Winter time ozone mixing ratios are on the order of 4 ppmv at 50 hPa at 60°N. Thus, the LIMS error estimate in Table 1 of 0.29 ppmv amounts to only 7%, such that the ozone of the LIMS analysis case is being weighted about equally between the LIMS and the SBUV data. In the following three sections, the DAS ozone analyses are compared against independent data, to judge whether the assimilation of the LIMS ozone has led to improved ozone analyses.

3.2. Comparisons With SAGE I Profiles

[32] From Figure 1b, one can see that the SAGE measurements extend to 50°N and to 60°S during April. Total systematic error for SAGE I ozone is about 5% (0.2 to 0.3 ppmv) in the uppermost stratosphere [*McCormick et al*., 1984], but not including the effects of the small altitude adjustment of *Wang et al*. [1996]. The estimated ozone errors for LIMS are a bit larger and for SBUV at least twice as large. In the top two panels of Figure 8, we show the mean differences for the average ozone profiles for both the Control and the LIMS analysis cases minus the collocated SAGE I results for the Northern and the Southern Hemispheres. Results from the LIMS analysis agree with SAGE I better than do those from the Control for the upper stratosphere of the Southern Hemisphere. The mean differences for each case for the Northern Hemisphere are within ±0.5 ppmv and are not as significant. They are not significant in either hemisphere from about 10 to 200 hPa.

[33] Random ozone errors for individual SAGE I profiles in the low and middle stratosphere are on the order of 5% to 10%, increasing to 20% by 40 km and to as much as

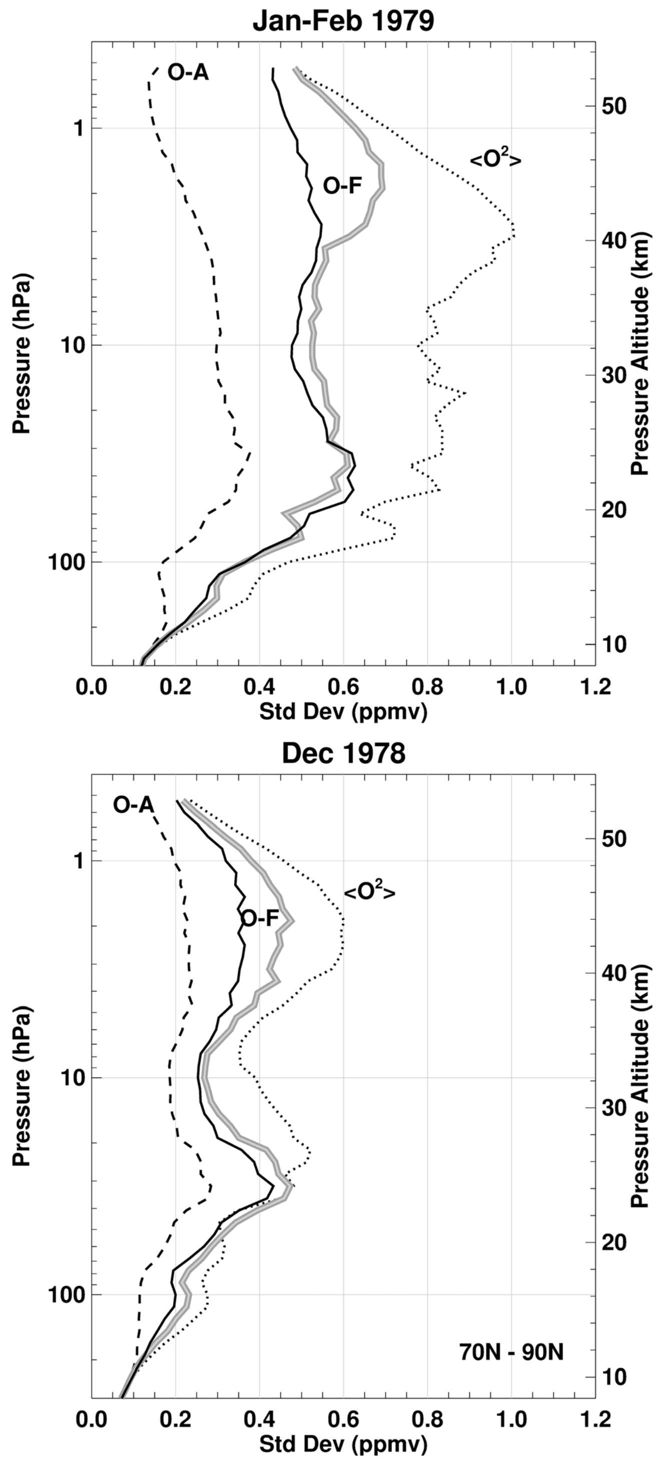

Figure 7. Profiles of the assimilation system standard deviation (SD) statistics for LIMS ozone (in ppmv) for (a) January and February 1979 and for 50°N to 70°N and (b) December 1978 and for 70°N to 90°N; observations minus forecast (O – F, solid black; O – F_{ctrl}, gray), observations minus analysis (O – A, dashed), and from the LIMS data (<O^2>, dotted).

40% by 50 km [*McCormick et al*., 1984]. The corresponding profiles of the RMS differences for the Control and for the LIMS analysis cases versus SAGE I are shown in the two bottom panels of Figure 8. RMS differences from both cases are only about 0.5 ppmv in the low to middle stratosphere for both hemispheres. However, for the comparisons in the Southern Hemisphere, the RMS differences for the Control increase to 1.0 ppmv near 1.5 hPa, reflecting the mean differences for the assimilated SBUV ozone versus SAGE I.

[34] Figure 9 shows the mean and RMS comparisons for the latter part of February 1979, or just after the launch of SAGE I. Now the differences for the Control

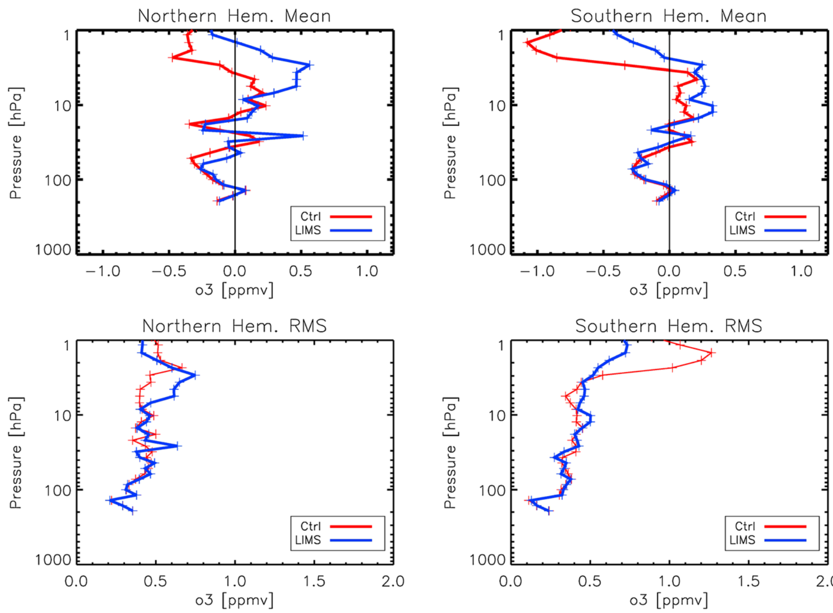

Figure 8. (top) Mean difference profiles from the Control and LIMS analysis cases minus SAGE I for April 1979; bias error for SAGE I is on the order of 0.3 ppmv in the upper stratosphere. (bottom) RMS difference profiles from the two cases. (left) Northern Hemisphere and (right) Southern Hemisphere.

exceed 1.0 ppmv near 1.5 hPa in the northern as opposed to the Southern Hemisphere. However, Northern Hemisphere comparisons with SAGE I were possible only between 50°N and 60°N for late February or in the region of the 1.0 ppmv differences from the LIMS analysis minus Control cases for that month (not shown). SAGE I comparison opportunities for March and May (not shown) are also weighted toward the middle and high northern latitudes. However, the mean differences for May are no larger than 0.7 ppmv in either hemisphere for either case. More noteworthy is that the RMS differences for the LIMS analysis ozone versus SAGE I indicate almost no changes between the two hemispheres for any of the 4 months, February through May.

[35] Based on the RMS comparisons in Figures 8 and 9 with the SAGE I ozone, it is concluded that the LIMS ozone provides some improvement for analyses of upper stratospheric ozone. Further, the hemispheric mean and RMS differences for the Control results indicate that there are slight biases in the SBUV ozone at those altitudes. *Bhartia et al.* [2012] report that the vertical resolution for SBUV v8.6 ozone is degraded somewhat in the uppermost stratosphere for measurements and retrievals at high solar zenith

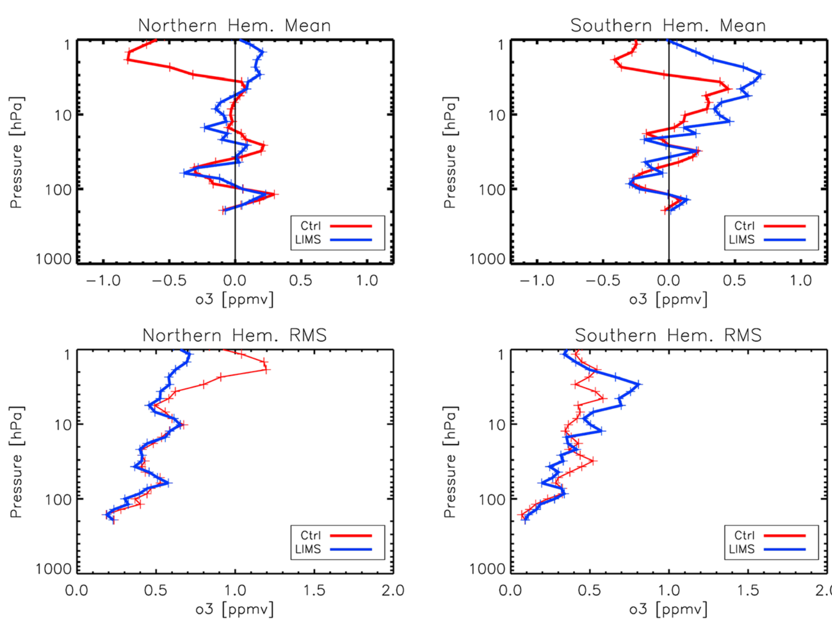

Figure 9. As in Figure 8 but for February 1979.

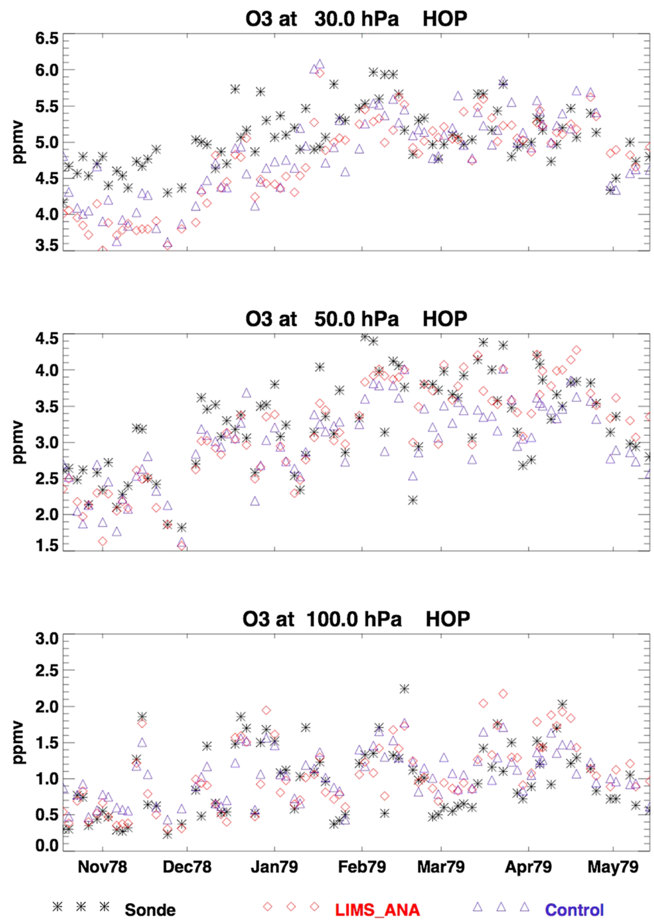

Figure 10. Time series of the ozonesonde data at 30, 50, and 100 hPa for Hohenpeissenberg with corresponding results from the Control and the LIMS analysis for November 1978 to May 1979 (abscissa has tics denoting the middle of each month). Systematic sonde errors are on the order of 0.25 ppmv at 30 hPa, 0.15 ppmv at 50 hPa, and 0.05 ppmv at 100 hPa. Sonde (asterisks); LIMS analysis (diamonds); Control (triangles).

angles, and that effect may explain the pattern of increasing differences toward 60°S and toward 80°N for April (Figure 3), at about 40°N to 50°N for November and December (Figures 4 and 5), and at 40°N for January (Figure 6). The vertical resolution (~3.7 km) of LIMS ozone is similar to that from SAGE I for the upper stratosphere, so they ought to be measuring the vertical ozone gradients faithfully. Since LIMS measured the atmospheric radiance from the Earth's limb and for both day and night, its ozone profiles have no dependence on solar zenith angle.

3.3. Comparisons With Ozonesonde Profiles at Hohenpeissenberg and at Wallops Island

[36] Next, we evaluate the quality of the lower stratospheric ozone from the LIMS analysis and Control cases based on comparisons with ozonesonde profiles at two station locations. Each analysis profile is the one closest in longitude, latitude, and time to the sonde ascent location. Figure 10 shows time series of the ozonesonde data at 30, 50, and 100 hPa for Hohenpeissenberg, Germany (HOP at 47.8°N, 11.0°E, taken about every other day) and the colocated results from the Control and LIMS analysis cases for the period of November 1978 through May 1979. Sondes at HOP measure ozone using the Brewer-Mast (BM) bubbler technique, and their data were accessed from the World Ozone and Ultraviolet Data Center. Data precision is 3% to 5%, and accuracy is about 5% in the stratosphere up to about 30 km [*Smit and Kley*, 1998]. The root-sum-square (RSS) sonde error is 6% to 7%.

[37] Mean differences (analysis minus sonde) and RMS differences for the set of sonde comparisons at HOP are given for the Ctr and the ANA cases in Table 3 in terms of absolute ozone values. Systematic BM sonde errors are on the order of 0.25 ppmv at 30 hPa, 0.15 ppmv at 50 hPa, and 0.05 ppmv at 100 hPa. At 30 hPa, the mean differences are −0.22 and −0.31 ppmv, respectively. The corresponding RMS differences from each case are about 0.47 ppmv (~10%) and larger than the root-sum-square (RSS) sonde error, but they are on the order of the combined satellite and sonde errors. Further, because the GEOS results are being compared essentially with point measurements, the RMS ozone differences have an extra component due to the differing spatial scales for the model versus the measurements. At 50 hPa, the mean differences are also negative or about −0.24 ppmv for the Ctr and −0.09 ppmv for the ANA case; the mean difference for the Ctr is significantly

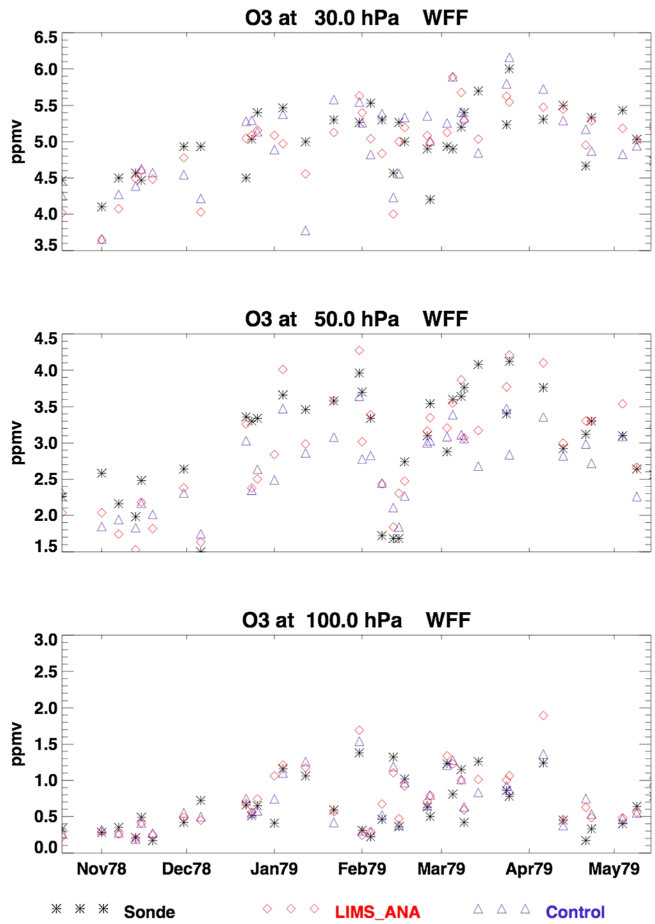

Figure 11. As in Figure 10 but for the ozonesonde data at Wallops Island.

negative compared with the sonde error of 0.15 ppmv. RMS values are 0.32 to 0.36 ppmv (~14%). At 100 hPa, the mean differences are slightly positive (0.09 ppmv) and not significant, and the RMS values are 0.29 ppmv.

[38] Figure 11 is analogous to Figure 10, but for the electrochemical concentration cell (ECC) ozonesonde measurements at Wallops Island, Virginia (WFF at 37.9°N, 284.5°E), where launches were made approximately weekly and often in conjunction with the expected overpass of NIMBUS 7. ECC sondes have a precision of 3–4% and an accuracy of 4–5%, or equivalent to the values for the BM sondes. Particularly noteworthy from Figure 11 is that both the Ctr and the ANA results are tracking the short-period variation of the sonde ozone at 50 hPa in late February, a feature related to the splitting of the polar vortex following the final winter warming of 1979. The mean differences in Table 3 at WFF are effectively zero at 30 hPa. RMS differences at WFF are larger than those at HOP or about 0.65 ppmv, but then the number of pairings at WFF is only about one third of that for the HOP sample. At 50 hPa the mean differences for the Ctr are biased negative and are significant (−0.25 ppmv), while the ANA results are near zero. Yet the RMS values at this level are near 0.51 ppmv from both cases, indicating that differences versus the sonde are mostly due to

Table 3. Mean and RMS Differences of the Correlative Ozone Versus the Control (Ctr) and the LIMS Analysis (ANA) Ozone, Based On the Ozonesonde Results at Hohenpeissenberg (HOP-Top) and at Wallops Island (WFF-Bottom)[a]

Pressure (hPa)	Ctr-Sonde (ppmv)	LIMS(ANA)-Sonde (ppmv)	RMS diff, Ctr-Sonde (ppmv)	RMS diff, LIMS(ANA)-Sonde (ppmv)
		Hohenpeissenberg		
30	−0.224	−0.312	0.468	0.479
50	−0.242	−0.089	0.319	0.361
100	0.090	0.089	0.293	0.289
		Wallops Island		
30	0.035	0.006	0.675	0.624
50	−0.252	−0.008	0.518	0.496
100	0.050	0.093	0.186	0.214

[a]A negative mean difference indicates that sonde ozone is higher than analyzed ozone. Systematic sonde errors are on the order of 0.25 ppmv at 30 hPa, 0.15 ppmv at 50 hPa, and 0.05 ppmv at 100 hPa.

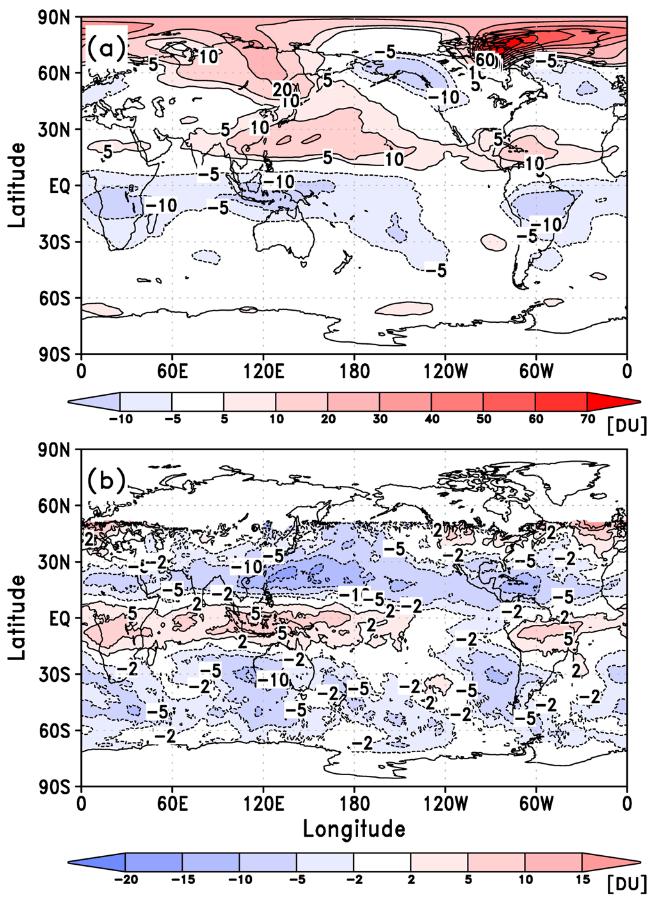

Figure 12. (a) Monthly mean, total ozone difference for January, LIMS analysis minus control (in DU); and (b) monthly mean, RMS difference versus TOMS from the two cases in terms of the RMS (LIMS anal – TOMS) – RMS (Ctrl – TOMS). Negative (blue) values indicate a better agreement from the LIMS analysis cases versus TOMS.

the disparity between their spatial scales. At 100 hPa, there is little to no mean difference in the two results, and the RMS values are on the order of 0.2 ppmv. In summary, there is a significant improvement from the assimilation of the LIMS ozone at both HOP and WFF at 50 hPa, but not at 30 and 100 hPa.

[39] One curiosity is that there is a persistent negative difference for both the Control and the LIMS analysis results versus the ozonesonde in winter at 30 hPa at HOP, but not at WFF (cf. Figures 10 and 11). *Logan* [1994] notes that the BM sonde technique tends to underestimate ozone, due to decreases in its flow pump efficiencies at the higher altitudes. However, the average correction factor (CF) needed to match the HOP ozone profiles to colocated Dobson total ozone measurements is only a bit larger (or 1.13) for the November 1978 through February 1979 sondes than for the March and April 1979 soundings (or 1.09). Furthermore, *Logan* [1994] showed that for HOP, the CF value is about 1.10 and was very stable for the entire period of 1970 to 1994. Therefore, it is considered more likely that the differences for the winter months in Figure 10 are due to a slight bias for the SBUV ozone at 30 hPa, particularly because the results from the LIMS analyses at this level are weighted toward the SBUV data and its small estimated error (~4%). Measurements at HOP during winter often occur near the edge of the polar vortex, where transport and/or mixing affect ozone but are not resolved in altitude accurately by SBUV. Therefore, it is postulated that there is a slight wintertime bias for the SBUV ozone at 30 hPa at HOP—similar to the feature associated with the QBO wind transition of the tropical latitudes (see section 3.1).

[40] One other finding is worthy of mention. The LIMS v6 algorithm does not consider the effects of horizontal gradients in ozone along the tangent-layer view path [*Remsberg et al.*, 2007, Figure 5a]. We found that this omission explains a systematic difference of up to 10% in the retrieved LIMS ozone profiles for the opposing viewing directions of its ascending versus its descending orbital segments. At 30 to 100 hPa, the ascending ozone is smaller than the descending ozone in the Northern Hemisphere and notably near the edge of polar vortex. This circumstance occurs because the LIMS sensor is viewing across the larger ozone gradients at the edge of the vortex; that bias effect is not included in the estimates of the LIMS ozone profile uncertainties in Table 2. Even so, such gradient effects do not explain the apparent, wintertime bias at 30 hPa in Figure 10 between the Control (SBUV) and the HOP ozone.

3.4. Comparisons With TOMS Total Ozone

[41] Since the retrieval of SBUV v8.6 partial ozone columns is not anchored by its associated total ozone measurement [*Bhartia et al.*, 2012], it is appropriate to compare the assimilated results of this study with the

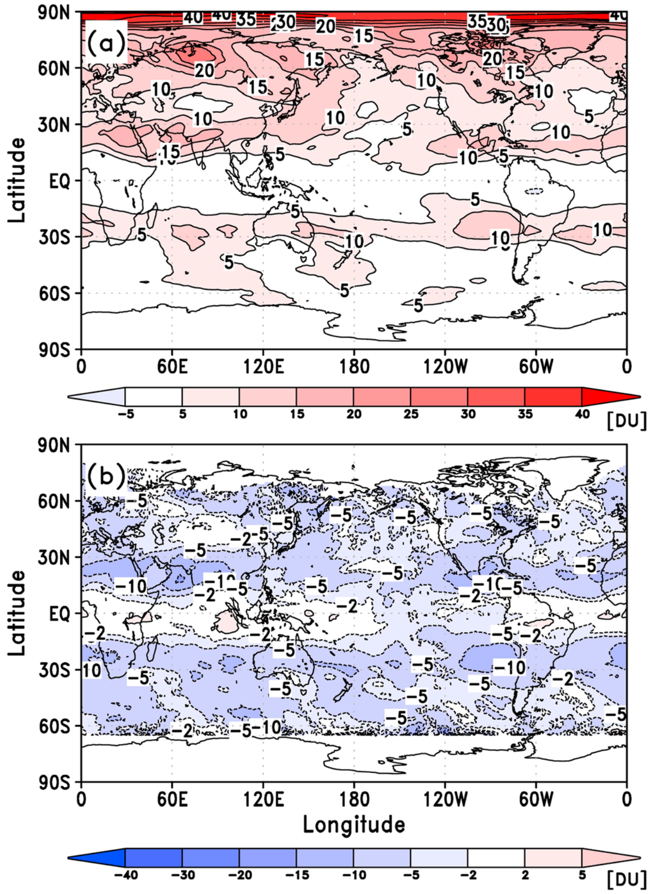

Figure 13. As in Figure 12 but for April.

independent total ozone distributions from TOMS v8. *Labow et al*. [2004] compared TOMS v8 ozone with total ozone from ground stations and reported that the TOMS values were high in 1978–1979, but by no more than 1% to 2%. On the other hand, they also reported that the TOMS ozone values of less than 275 DU (in the tropics) are likely too low by about 1% to 2% due to a bias from the 225 DU climatology used in the TOMS retrieval algorithm.

[42] At the outset, we point out that total column ozone from the Control case is smaller than the TOMS total ozone and by at least 20 DU nearly everywhere (not shown). This result is consistent with our finding of a negative bias in Table 3 for Ctr versus sonde at 50 hPa and indicates that the SBUV v8.6 ozone may be underestimated for the layer from 63 to 40 hPa. There may also be a bias due to the model tropospheric partial column ozone, especially since neither the Control nor the LIMS cases are assimilating measured total ozone values, too. Instead, we first show in Figure 12a (at top) the monthly averaged differences for total column ozone (in DU) from the two cases, or LIMS analysis minus Control, for January 1979. At the middle to high latitudes of the Northern Hemisphere, the differences are large and oscillate from positive (red, 10 to 20 DU or more) to negative (blue, 5 to 10 DU) and with a clear, zonal wave-1 or wave-2 pattern. At about 20°N and near the pole, the average differences are positive at all longitudes. Thus, the differences in the Northern Hemisphere strongly suggest that the assimilation of the LIMS ozone is adding information about the transport of ozone within the lower and middle stratosphere. At middle latitudes of the Southern Hemisphere, the differences are uniformly less than 5 DU for this summer month, when zonal wave activity is weak; the assimilation of LIMS ozone offers little improvement there. In the equatorial region, the differences are negative everywhere, except near 90°W, and there are indications that the negative differences are largest (−10 DU) over South America, Africa, and Indonesia. Part of that negative bias is reflective of the differences in the tropics for the LIMS versus the Control (SBUV) cases shown in Figure 6 at about 10 to 20 hPa. In addition, it is possible that the LIMS ozone values are biased low for several kilometers above the tropical tropopause, but not low enough that they were removed prior to the assimilation process (see Appendix A). Even though the assimilated ozone is weighted toward SBUV in that region, the LIMS ozone is still making a contribution. Just why the LIMS assimilation case might lead to ozone minima over preferred geographical regions is unclear, but it is common to see organized convective systems and their associated lower values of upper tropospheric ozone over tropical land areas.

[43] Figure 12b (at bottom) shows the January average distribution of the RMS differences between the LIMS analysis and the Control cases versus the colocated TOMS ozone. Specifically, the plot shows the RMS (LIMS anal − TOMS) − RMS (Ctrl − TOMS). Negative (blue) values indicate that there is better agreement (or smaller

RMS values) from the LIMS analysis versus TOMS ozone differences, and that finding extends to most locations outside the tropics. The positive (red) RMS differences for the tropics are on the order of 5 DU or less (< 2%), where there may be a contribution from a residual low bias in the LIMS ozone of the lower tropical stratosphere.

[44] Figure 13 is analogous to Figure 12 but for April 1979. Figure 13a shows that total column ozone values from the LIMS analysis case are greater than from the Control at nearly all latitudes. In the tropics, the column differences are near zero and in agreement with the finding of little to no ozone differences in the middle stratosphere in Figure 3. The positive differences at northern midlatitudes are now nearly uniform with longitude and consistent with the fact that most of the meridional exchange and wave-induced mixing of the ozone had occurred by April [e.g., *Leovy et al.*, 1985]. Figure 13b shows negative (blue) differences nearly everywhere, even in the tropics, and again indicates that there is better agreement between the LIMS analysis and the TOMS ozone. In summary and except for the tropics in January, it is clear from these comparisons with TOMS that the assimilation of the LIMS ozone is providing quantitative improvements to the stratospheric ozone analyses for both January and April, and most likely throughout the entire LIMS period of 1978–1979.

4. Summary and Conclusions

[45] We have assimilated ozone from the LIMS v6 data set along with that from SBUV v8.6 for a LIMS analysis study with the GEOS-5 v7.2 model and its data assimilation system for the period November 1978 through May 1979. This study makes use of an updated module to account for the chemical loss of ozone due to the reactive chlorine levels that were present during the time period of LIMS. A Control case was conducted for comparison, wherein only SBUV data were assimilated. LIMS ozone complements the SBUV data by providing measurements in darkness, as well as in daylight regions, and it provides profile data of a higher vertical resolution.

[46] We find that the LIMS ozone provides improved results at the high latitudes in winter/spring and in the polar night region, where SBUV data are lacking. We also find slight increases in the zonally averaged ozone at middle to high latitudes of the uppermost stratosphere. Separate comparisons with SAGE I observations indicate that the LIMS analysis ozone agrees better with them in the upper stratosphere. The inclusion of the LIMS ozone also improves the analysis ozone distributions in the lower stratosphere at subtropical and middle latitudes, based on comparisons of total column ozone with TOMS ozone. This finding is consistent with the nearly equal uncertainties and contributions of the SBUV v8.6 and the LIMS v6 ozone profiles. In addition, the LIMS ozone resolves some coherent vertical structure in the ozone at low latitudes that is associated with a phase transition for the QBO winds. It is concluded that the addition of ozone profiles from limb sounders can improve future ozone reanalyses, especially if their associated measurement errors are comparable to those from the present operational ozone sounders.

[47] The good quality of the lower stratospheric analyses from both the LIMS and the SBUV ozone is supported by independent comparisons with time series of ozonesonde data at HOP and at WFF, although there are indications that the analysis ozone is underestimated at 50 hPa for the Control case. Further, the LIMS assimilation studies have been helpful with validating the LIMS v6 ozone, especially during this early time period when there were relatively few correlative ozone measurements. In particular, we found that the presence of LIMS ozone values having a modal value of 0.05 ppmv in the lower tropical stratosphere led to a low bias in its analysis results at the adjacent, higher altitudes. Researchers are urged to consider whether those low values could also affect their studies with the LIMS v6 ozone. We conclude that data assimilation is a powerful tool for evaluating the quality of satellite ozone data sets and that v7.2 of the GEOS-5 model can improve ozone analyses in 1978–1979 by the assimilation of the LIMS ozone.

Appendix A: Excessively Low Tropical Ozone Values in the LIMS V6 Data Set

[48] We discovered from our preliminary LIMS analyses with MERRA that the ozone results were significantly affected by the presence of many small values in the LIMS v6 ozone data (≃0.05 ppmv) of the lower stratosphere at tropical to middle latitudes. As an example, Figure A1 shows results at the equator for 6 January 1979. The zonal average LIMS analysis ozone at the 40 hPa level and below is noticeably smaller than from an initial Control case that used SBUV v8.0 ozone. A minimum average value of about 0.03 ppmv was obtained near 100 hPa from those LIMS analyses, which is excessively low ozone for the tropical lower stratosphere. That preliminary study also employed percentage values for the LIMS error vector from *Remsberg et al*. [2007], giving undue significance to very small values. Furthermore, the average January ozone shown in Figure A1 from the LIMS profiles that were ingested is also much greater than the results from the LIMS analyses.

[49] Data assimilation normally assumes that the observation (O) and forecast (F) errors are Gaussian, such that the (O − F) residual is also Gaussian [e.g., see *Struthers et al*., 2002; *Lahoz et al*., 2007; *Stajner et al*., 2001]. Conversely, if (O − F) does not have a Gaussian error distribution, at least one of O or F does not have Gaussian errors. Figure A2a shows the frequency distribution of LIMS v6 ozone between 30°S and 30°N and 70–100 hPa for 25 October 1978—the first day of LIMS data. It shows a reasonable, relatively flat distribution of points ranging between 0.1 and 1 ppmv. There is also a second concentration of points with a mode centered near 0.05 ppmv. The frequency distribution of the (O − F) values is in Figure A2b and should be centered at zero. However, it is strongly skewed, with a negative peak frequency of −0.05 ppmv and a tail extending to 0.8 ppmv. The GSI package gives an observation minus analysis (O − A) distribution that is even more skewed to the right, although centered at 0.0 ppmv. The LIMS ozone value of 0.05 ppmv is equivalent to the effect of the measured radiance noise for tangent layer ozone in the lower stratosphere. Therefore, we screened out ozone values less than 0.1 ppmv and for pressure levels greater than 50 hPa from consideration for our final LIMS analysis run. The resultant

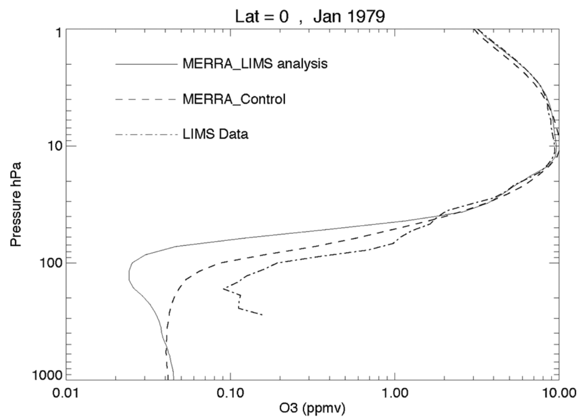

Figure A1. Zonally averaged ozone at the equator from the preliminary Control and the LIMS analysis cases for 6 January 1979 as compared with January average LIMS v6 ozone.

O – F and O – A distributions are shown in Figure A2c and are more nearly Gaussian. The total number of points in Figure A2c is reduced accordingly (cf. the vertical scales between Figure A2b and Figure A2c).

[50] Figure A3 shows zonal average LIMS ozone for 6 January 1979, both prior to and after being screened of the small noise-limiting values. The sharp reversal to larger values in the upper troposphere in the original data is likely due to residual emission from subvisible cirrus that was not already flagged by the cloud detection algorithm applied to the generation of the archived v6 data set. Our screened profile extends only to 100 hPa and has a minimum value near 0.4 ppmv. As indicated in Figures 3 through 6, there is little difference between the tropical ozone distributions of the lower stratosphere from the final Control and the LIMS analyses because the analysis ozone in that region is weighted toward the SBUV data.

[51] The appearance of unrealistically low ozone values in the lower stratosphere in LIMS v6 is a result of the retrieval

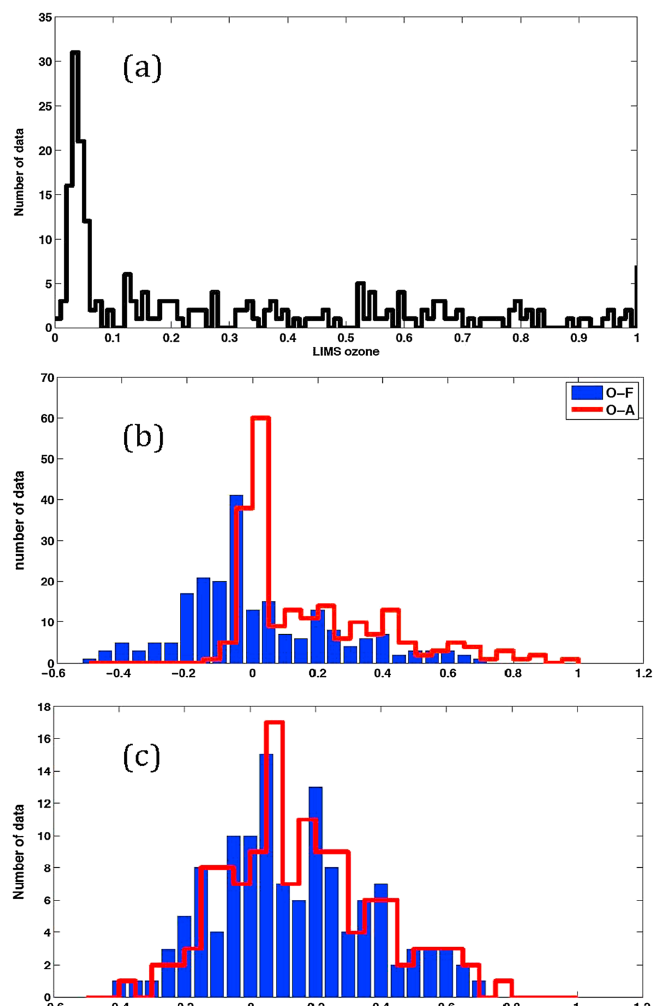

Figure A2. (a) Frequency distribution of LIMS ozone between 30°S and 30°N, 70 to 100 hPa, for 25 October 1979 that include excessively low values of ≤0.05 ppmv; (b) resultant frequency distribution of observation minus forecast ((O – F) values (blue filled) and observation minus analysis (O – A) values (red) provided with the GSI package, both in intervals of 0.05 ppmv; (c) as in Figure A2b, but the corresponding O – F and O – A distributions after removal of the excessively low values.

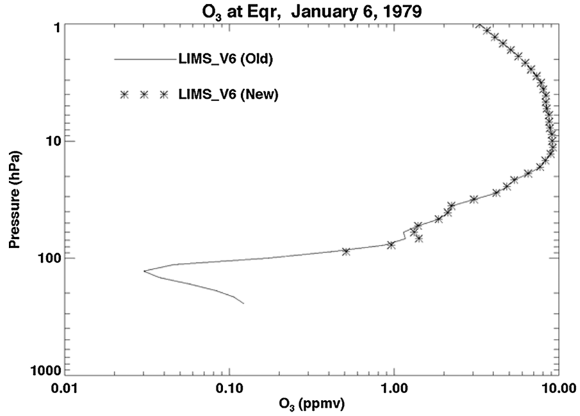

Figure A3. Average LIMS v6 ozone at the equator for 6 January 1979, both prior to (old) and after screening (new) for values less than 0.1 ppmv.

method, known bias errors, and the characteristics of the broadband LIMS ozone channel itself. The LIMS v6 retrieval uses an onion-peeling approach with an emissivity growth approximation (or EGA) for calculations of the tangent layer, line-of-sight (LOS) emissivity in the LIMS forward model [*Remsberg et al.*, 2004]. The method is sensitive to errors for the tropical lower stratosphere, where the ozone mixing ratio is small, the vertical ozone gradient is large, and the systematic errors in the retrieved temperatures or from the ozone forward model are significant. *Remsberg et al.* [2007, Table 1] estimate that biases in the radiances from the forward model of LIMS ozone can lead to errors of ~15% in ozone mixing ratio at 100 hPa at midlatitudes and up to 30% at tropical latitudes. Those estimates were obtained by comparing ozone retrieved using the LIMS v6 emissivity tables with results of line-by-line calculations. Underestimates of 15% to 30% can lead to retrieved ozone approaching zero well above the 100 hPa level. Due to the convergence logic of the retrieval algorithm, the ozone does not go to zero but rather to noise equivalent radiance (NER) values of about 0.05 ppmv. The spectrally wide, LIMS ozone channel is optically thin outside the region of the v_3 and v_1 ozone bands and observes atmospheric radiances from cloud tops and the upper troposphere. Thus, the LIMS ozone retrieval can proceed on to the next lower tangent layer and converge once again to the NER. This circumstance explains the rather large number of points in Figure A2a having modal values near 0.05 ppmv.

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